

# The Seebeck coefficient in correlated low dimensional organic metals

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We study the influence of inelastic electron-electron scattering on the temperature variation of the Seebeck coefficient in the normal phase of quasi-one-dimensional organic superconductors. The theory is based on the numerical solution of the semi-classical Boltzmann equation for which the collision integral equation is solved with the aid of the electronic umklapp scattering vertex calculated by the renormalization group method. We show that the one-loop renormalization group flow of momentum and temperature dependent umklapp scattering, in the presence of nesting alterations of the Fermi surface, introduce electron-hole asymmetry in the energy dependence of the anisotropic scattering time. This is responsible for the enhancement of the Seebeck coefficient with respect to the band  $T$ -linear prediction and even its sign reversal around the quantum critical point of the phase diagram, namely where the interplay between antiferromagnetism and superconductivity along with the strength of spin fluctuations are the strongest. Comparison of the results with available data on low dimensional organic superconductors is presented and critically discussed.

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## I. INTRODUCTION

In the past few years we have seen expanding interest in the Seebeck coefficient as a sensitive probe of fluctuations encased in the quantum critical behaviour of correlated electrons. This has been exemplified both experimentally and theoretically for quantum critical points in heavy fermions<sup>1–5</sup>, pnictides<sup>6</sup>, and for hole<sup>7–10</sup>, and electron-doped<sup>11,12</sup> cuprates. In organic superconductors like the Bechgaard salts (TMTSF)<sub>2</sub>X series, also known to exhibit quantum criticality, the measurements of the Seebeck coefficient have been the subject of numerous reports following their discovery<sup>13–19</sup>, and this, until very recently<sup>20</sup>. However, these works have found very little theoretical echo as to the possible part played by quantum fluctuations in the thermoelectric response seen in these correlated quasi-one dimensional (quasi-1D) metals. This topic that has remained essentially unexplored so far<sup>21</sup>, will be the main focuss of the present work.

The quantum critical behaviour of the Bechgaard salts is known to result from the juncture of a declining spin-density-wave (SDW) state with the onset of a superconducting (SC) dome under pressure<sup>22–26</sup>. The signatures of quantum criticality have been chiefly revealed by the observation of linear- $T$  resistivity<sup>27</sup>, whose strength scales with the distance from the quantum critical point (QCP) along the pressure axis. Among other fingerprints of quantum criticality, linear resistivity was also found to scale with the amplitude of SDW fluctuations seen by NMR and with the size of the critical temperature  $T_c$  for superconductivity<sup>25,28–30</sup>.

The contributions of the renormalization group (RG) approach to the understanding of these quantum critical features have been the purpose of several works in the past<sup>31–33</sup>. In the framework of the quasi-1D electron gas model for instance, that is how the characteristic sequence of instabilities lines and the scaling of spin

fluctuations with the size of  $T_c$  could be fairly well simulated when the antinesting amplitude of the quasi-1D electron band structure is used as a tuning parameter for the QCP<sup>33–35</sup>.

More recently, the RG calculations for the umklapp vertex was shown to serve as an input to the linearized Boltzmann theory of electrical transport<sup>36</sup>. From this combination of techniques, first proposed by Buhmann *et al.*<sup>9</sup> in the context of the 2D Hubbard model for the cuprates, the metallic resistivity across the QCP could be calculated along the pressure - antinesting - axis and the results congruently compared with experiments in the Bechgaard salts<sup>27</sup>.

In this work we further exploit the RG-Boltzmann approach and derive the Seebeck coefficient for correlated quasi-1D metals. The numerical integration of the linearized Boltzmann equation when fed in by the RG umklapp vertex function, allows a microscopic determination of the energy variation of the anisotropic electron-electron scattering time across the Fermi surface. This variation is mostly influenced by SDW fluctuations and is anisotropic on the Fermi surface. It introduces deviations with respect to the Seebeck coefficient obtained in the band limit, which is positive and linear in temperature for hole carriers in materials like the Bechgaard salts. The deviations take the form of enhancements that can be not only positive, but also negative or electron like in character. The latter can lead to the sign reversal of the Seebeck coefficient, especially in the neighborhood of the QCP where the interplay between SDW and SC, together with the amplitude of the SDW fluctuations in the metallic state, are the strongest. These results offer an interesting avenue for the understanding of the sign reversal in the Seebeck coefficient of the Bechgaard salts near their QCP.

The theory is broadened to systems with stronger umklapp scattering that favours a Mott instability in the 1D

portion of their metallic state, which can be approached by the weak coupling RG from the high temperature domain. The results are confronted to the measurements of the Seebeck coefficient for prototype members of the sulfur based compounds, the (TMTTF)<sub>2</sub>X, known as the Fabre salts series<sup>37</sup>. These compounds are characterized by a more pronounced dimerization of the organic stacks which magnifies umklapp scattering and favours a crossover toward a 1D Mott insulating state.

In Sec. II, we use the linearized Boltzmann theory to derive the expression of the Seebeck coefficient for a quasi-1D three-quarter filled hole band of a lattice of weakly dimerized chains. In Sec. III, the momentum-resolved renormalized umklapp vertex entering the Boltzmann equation is computed by the RG, in the conditions of the quasi-1D electron gas model simulating the sequence of instabilities found in the Bechgaard salts. In Sec. IV, we present the temperature variations of the Seebeck coefficient across the quantum critical point of the model and examine their link with the energy profile of the inelastic scattering time. In Sec. V, a comparison of the results is made with available data for (TMTSF)<sub>2</sub>X, and on a broader basis for the more correlated compounds (TMTTF)<sub>2</sub>X. We conclude in Sec. VI.

## II. LINEARIZED BOLTZMANN THEORY OF THE SEEBECK COEFFICIENT

We consider the semiclassical Boltzmann equation for the variation of the quasi-particle Fermi distribution function  $f$  in the presence of collisions and a thermal gradient  $\nabla_{\mathbf{r}}T$ . In steady-state conditions, it takes the form

$$\left[ \frac{\partial f(\mathbf{k})}{\partial t} \right]_{\text{coll}} = e \mathcal{E} \cdot \nabla_{\mathbf{k}} f - \frac{(E_{\mathbf{k}} - \mu)}{T} \nabla_{\mathbf{r}} T \cdot \nabla_{\mathbf{k}} f, \quad (1)$$

where  $E_{\mathbf{k}}$  is the carrier spectrum,  $\mu$  the chemical potential,  $e$  the electron charge and  $\mathcal{E}$  the electric field set up by the thermal gradient. The collision integral for an array of  $N_P$  chains of length  $L$  takes the form

$$\begin{aligned} \left[ \frac{\partial f(\mathbf{k})}{\partial t} \right]_{\text{coll}} = & -(LN_P)^{-2} \sum_{\mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4} \frac{1}{2} |\langle \mathbf{k}, \mathbf{k}_2 | g_3 | \mathbf{k}_3, \mathbf{k}_4 \rangle| \\ & - \langle \mathbf{k}, \mathbf{k}_2 | g_3 | \mathbf{k}_4, \mathbf{k}_3 \rangle|^2 \frac{2\pi}{\hbar} \delta_{\mathbf{k}+\mathbf{k}_2, \mathbf{k}_3+\mathbf{k}_4 \pm \mathbf{G}} \\ & \delta(E_{\mathbf{k}} + E_{\mathbf{k}_2} - E_{\mathbf{k}_3} - E_{\mathbf{k}_4}) \\ & \times \{ f(\mathbf{k}) f(\mathbf{k}_2) [1 - f(\mathbf{k}_3)] [1 - f(\mathbf{k}_4)] \\ & - [1 - f(\mathbf{k})] [1 - f(\mathbf{k}_2)] f(\mathbf{k}_3) f(\mathbf{k}_4) \}, \end{aligned} \quad (2)$$

From the Fermi Golden rule, the transition probability per unit of time is given by the matrix element  $\langle \mathbf{k}, \mathbf{k}_2 | g_3 | \mathbf{k}_3, \mathbf{k}_4 \rangle$  for longitudinal umklapp processes where  $\mathbf{G} = (4k_F, 0)$  is the longitudinal reciprocal lattice wave vector and  $k_F$  is the 1D Fermi wavevector.

We proceed to the linearization of the Boltzmann equation by introducing<sup>38</sup>

$$f(\mathbf{k}) = \frac{1}{e^{\beta(E_{\mathbf{k}} - \mu) - \phi_{\mathbf{k}}} + 1}, \quad (3)$$

where  $\phi_{\mathbf{k}}$  is a normalized deviation to thermal equilibrium and  $\beta = 1/k_B T$ . In the tight-binding approximation, the hole band spectrum for a linear array of  $N_P$  weakly coupled dimerized chains is given by

$$E_{\mathbf{k}} = \sqrt{2(t^2 + \delta t^2) + 2(t^2 - \delta t^2) \cos ka} + \epsilon_{\perp}(k_{\perp}), \quad (4)$$

where  $t \pm \delta t$  are the transfer integrals within and between the dimers ( $\delta t > 0, \delta t \ll t$ ). Here  $a$  is the lattice spacing along the chains, namely the distance between dimers. The transverse part of the hole spectrum is given by

$$\epsilon_{\perp}(k_{\perp}) = 2t_{\perp} \cos k_{\perp} d_{\perp} + 2t'_{\perp} \cos 2k_{\perp} d_{\perp}, \quad (5)$$

where  $t_{\perp}$  and  $t'_{\perp}$  are the first and the second-nearest neighbour transfer integrals in the direction perpendicular to the chains.

For small deviations with respect to equilibrium, the Fermi distribution becomes

$$f(\mathbf{k}) \simeq f^0(\mathbf{k}) + f^0(\mathbf{k})[1 - f^0(\mathbf{k})]\phi_{\mathbf{k}}, \quad (6)$$

where  $f^0(\mathbf{k})$  is the equilibrium distribution at  $\phi_{\mathbf{k}} = 0$ . Replacing (6) into Eqs (1) and (2) leads to the linearized Boltzmann equation

$$\begin{aligned} \mathcal{L}\phi_{\mathbf{k}} = & e\beta \mathcal{E} \cdot \mathbf{v}_{\mathbf{k}} - \beta^2 k_B (E_{\mathbf{k}} - \mu) \mathbf{v}_{\mathbf{k}} \cdot \nabla_{\mathbf{r}} T \\ \equiv & \mathcal{L}\phi_{\mathbf{k}}^{\mathcal{E}} - \mathcal{L}\phi_{\mathbf{k}}^T. \end{aligned} \quad (7)$$

The collision operator  $\mathcal{L}$  satisfies the integral equation

$$\mathcal{L}\phi_{\mathbf{k}}^j = \sum_{\mathbf{k}'} \mathcal{L}_{\mathbf{k}, \mathbf{k}'} \phi_{\mathbf{k}'}^j, \quad (j = \mathcal{E}, T). \quad (8)$$

where the kernel is given by

$$\begin{aligned} \mathcal{L}_{\mathbf{k}, \mathbf{k}'} = & (LN_P)^{-2} \sum_{\mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4} \frac{1}{2} |\langle \mathbf{k}, \mathbf{k}_2 | g_3 | \mathbf{k}_3, \mathbf{k}_4 \rangle| \\ & - \langle \mathbf{k}, \mathbf{k}_2 | g_3 | \mathbf{k}_4, \mathbf{k}_3 \rangle|^2 \frac{2\pi}{\hbar} \delta_{\mathbf{k}+\mathbf{k}_2, \mathbf{k}_3+\mathbf{k}_4 \pm \mathbf{G}} \\ & \delta(E_{\mathbf{k}} + E_{\mathbf{k}_2} - E_{\mathbf{k}_3} - E_{\mathbf{k}_4}) \\ & \times \frac{f^0(\mathbf{k}_2)[1 - f^0(\mathbf{k}_3)][1 - f^0(\mathbf{k}_4)]}{[1 - f^0(\mathbf{k})]} \\ & \times (\delta_{\mathbf{k}, \mathbf{k}'} + \delta_{\mathbf{k}_2, \mathbf{k}'} - \delta_{\mathbf{k}_3, \mathbf{k}'} - \delta_{\mathbf{k}_4, \mathbf{k}'}) \\ = & \sum_{i=1}^4 \mathcal{L}_{\mathbf{k}, \mathbf{k}'}^{[i]}, \end{aligned} \quad (9)$$

and which can be written as the sum of four contributions. The explicit expressions for the diagonal ( $\mathcal{L}^{[1]}$ ) and off-diagonal ( $\mathcal{L}^{[2-4]}$ ) terms are calculated according to Ref.<sup>36</sup>, in the limit of the quasi-1D electron gas model. Their expressions given in Appendix A are generalizations at arbitrary energy distance from the Fermi level.

The electric current density along the chains resulting from a longitudinal thermal gradient  $\nabla_a T$  and the induced electric field  $\mathcal{E}_a$  in leading order is given by

$$j_a = \frac{2e}{LN_\perp d_\perp} \sum_{\mathbf{k}} v_{\mathbf{k}}^a f(\mathbf{k})$$

$$\simeq \frac{2e}{LN_\perp d_\perp} \sum_{\mathbf{k}} v_{\mathbf{k}}^a f^0(\mathbf{k}) [1 - f^0(\mathbf{k})] (\phi_{\mathbf{k}}^\mathcal{E} - \phi_{\mathbf{k}}^T), \quad (10)$$

where  $v_{\mathbf{k}}^a$  is the carrier velocity along the  $a$  direction. Introducing the normalized deviations  $\bar{\phi}_{\mathbf{k}}^\mathcal{E} = \phi_{\mathbf{k}}^\mathcal{E} / (e\beta v_{\mathbf{k}_F}^a \mathcal{E}_a)$  and  $\bar{\phi}_{\mathbf{k}}^T = \phi_{\mathbf{k}}^T / [\beta^2 k_B v_{\mathbf{k}_F}^a (E_{\mathbf{k}} - \mu) \nabla_a T]$ , which have the units of time, this expression can be recast in the form

$$j_a = K_{11} \mathcal{E}_a - K_{12} \nabla_a T, \quad (11)$$

which leads in the absence of charge current ( $j_a = 0$ ) to the expression of longitudinal Seebeck coefficient  $Q_a$ , as the ratio

$$Q_a = \frac{\mathcal{E}_a}{\nabla_a T} = \frac{K_{12}}{K_{11}}. \quad (12)$$

Since the product  $f^0[1 - f^0]$  is strongly peaked at the Fermi level  $E_{\mathbf{k}} - \mu \equiv E = 0$ , a Sommerfeld expansion of the matrix elements  $K_{11}$  and  $K_{12}$  yields the following expression for the Seebeck coefficient,

$$Q_a = \frac{\pi^3 k_B^2 T}{3 |e|} \left\{ \left[ -\frac{d \ln \langle N(E, k_\perp) \rangle_{k_\perp}}{dE} - 2 \frac{d \ln \langle v_{E, k_\perp}^a \rangle_{k_\perp}}{dE} \right] - \frac{\partial \ln \langle \bar{\phi}_{E, k_\perp} \rangle_{k_\perp}}{\partial E} \right\}_{E=0}$$

$$= Q_a^0 + Q_a^c, \quad (13)$$

which can be separated into two contributions. The first, noted  $Q_a^0$ , is the sum of the two terms in brackets, which corresponds to the band contribution. It is linked to the energy derivatives of the density of states per spin,  $\langle N(E, k_\perp) \rangle_{k_\perp} = (\pi^{-1} \langle \partial k / \partial E_{\mathbf{k}} \rangle_{k_\perp})$ , and of the longitudinal velocity  $\langle v_{E, k_\perp}^a \rangle_{k_\perp} = (\hbar^{-1} \langle \partial E_{\mathbf{k}} / \partial k \rangle_{k_\perp})$ . Both quantities are averaged over the Fermi surface for a filling of one hole per dimer ( $\langle \dots \rangle_{k_\perp} = N_F^{-1} \sum_{k_\perp} \dots$ ). The second contribution,  $Q_a^c$ , is associated to collisions; it is proportional to the energy derivative  $\langle \bar{\phi}'_{E=0, k_\perp} \rangle_{k_\perp}$  averaged over the Fermi surface for the normalized deviations  $\bar{\phi}^\mathcal{E} = \bar{\phi}^T \equiv \bar{\phi}$ , namely the scattering time. The latter obeys the single integral equation

$$\mathcal{L} \bar{\phi}_{\mathbf{k}} = \sum_{i, \mathbf{k}'} \mathcal{L}_{\mathbf{k}, \mathbf{k}'}^{[i]} \bar{\phi}_{\mathbf{k}'} = 1, \quad (14)$$

whose explicit expression is given in (A6). Here  $\bar{\phi}_{\mathbf{k}} \rightarrow \bar{\phi}_{E, k_\perp}$  can be expressed as a function of the energy distance from the Fermi surface and the angle parametrized by  $k_\perp$ . The expression (13) is reminiscent of the Mott

formula for the Seebeck coefficient<sup>21,39</sup>. It should be stressed, however, that the scattering term results from the solution of the  $\mathbf{k}$  dependent integral equation (14), which goes beyond the relaxation time approximation used for the Mott result<sup>40</sup>.

### III. RENORMALIZED UMKLAPP VERTEX

#### A. The quasi-one-dimensional electron gas model

The temperature variation of the momentum dependent umklapp vertex part entering the collision operator of the Boltzmann equation (9) is calculated using the renormalization group technique in the framework of the quasi-one-dimensional electron gas model. In the model the longitudinal part of lattice model for the hole spectrum  $E_{\mathbf{k}}$  in (4) is linearized with respect to the 1D Fermi points  $\pm k_F$ . This gives

$$E_{\mathbf{k}} - \mu \approx \epsilon_{\mathbf{k}}^p = -\hbar v_F (pk - k_F) + \epsilon(k_\perp), \quad (15)$$

where  $p = \pm$  refers to right and left moving carriers along the chains and  $v_F = (t^2 - \delta t^2)a / (\hbar \sqrt{2t^2 + 2\delta t^2})$  is the longitudinal Fermi velocity. According to band calculations, the hopping integrals will be fixed at  $t/k_B = 2700\text{K}$  and  $t_\perp/k_B = 200\text{K}$  as typical figures for hopping integrals in compounds like the Bechgaard salts. A second harmonic is added to the transverse tight-binding spectrum which acts as an anti-nesting tuning parameter  $t'_\perp \ll t_\perp$ . Anti-nesting is considered as the main parameter simulating the pressure in the model.

Particles interact through three coupling constants defined on the warped Fermi surface sheets  $\mathbf{k}_F^p(k_\perp) = (k_F^p(k_\perp), k_\perp)$ , as parametrized by  $k_\perp$  from the condition  $\epsilon^p(\mathbf{k}_F^p) = 0$  (see the top panel of the Fig. 4). These are the backward and forward scattering amplitudes  $g_1(\mathbf{k}_{F,1}^-, \mathbf{k}_{F,2}^+, \mathbf{k}_{F,3}^-, \mathbf{k}_{F,4}^+)$  and  $g_2(\mathbf{k}_{F,1}^+, \mathbf{k}_{F,2}^-, \mathbf{k}_{F,3}^-, \mathbf{k}_{F,4}^+)$ , and the longitudinal umklapp scattering  $g_3(\mathbf{k}_{F,1}^p, \mathbf{k}_{F,2}^p; \mathbf{k}_{F,3}^p, \mathbf{k}_{F,4}^p)$ . All couplings are normalized by  $\hbar \pi v_F$  and develop from renormalization a momentum dependence on three independent transverse momentum variables.

We will follow previous works<sup>33,34,36</sup> and fix the bare initial repulsive values of the couplings consistently with different experiments and band calculations. Thus for the bare backward scattering, by taking  $g_1 \approx 0.32$ , one can reasonably account for the observed temperature dependent enhancement of uniform susceptibility<sup>41</sup>. For the bare longitudinal umklapp term  $g_3$ , its bare amplitude is non-zero but very weak, owing to the small dimerization of the organic stacks that introduces some half-filled character to the band. This yields to  $g_3 \approx g_1(2\delta t/t)$ , as a result of the modulation  $\delta t$  of longitudinal hopping integrals responsible for the dimerization gap<sup>42,43</sup>. According to band calculations at low pressure<sup>44</sup>,  $\delta t/t \approx 0.05 \dots 0.1$ , suggesting to take  $g_3 \approx 0.025$  in the following calculations. From these figures, the amplitude of the bare

forward scattering can be finally adjusted to the value  $g_2 \approx 0.64$ , in order to get from the low  $t'_\perp$  RG calculations (see Fig. 1) the right order of magnitude for the observed SDW scale, namely  $T_{\text{SDW}} \sim 10\text{K}$  for  $(\text{TMTSF})_2\text{X}$  at ambient pressure<sup>22</sup>. As a function of  $t'_\perp$ , the application of the RG generates a phase diagram compatible with the experimental situation<sup>22,27</sup>. There is nothing special in the above choice. Actually at small umklapp, it exists a whole range of reasonable coupling parameters that would yield a phase diagram comparable to Fig. 1 and then to similar results for the Seebeck coefficient.

As it will be discussed in more details in Sec. V A, one can extend the analysis to the more correlated sulfur based Fabre salts series  $(\text{TMTTF})_2\text{X}$  characterized by smaller band parameters and stronger umklapp scattering owing to a larger dimerization of the organic stacks.

### B. Renormalization group results

The RG approach to the above quasi-1D electron gas model has been described in detail in previous works<sup>32–36</sup>. In essence, it consists in the segmentation of infinitesimal energy shells on either side of the Fermi sheets into  $N_P$  patches, whose internal transverse momentum integration in the loop calculations, leads to as many  $k_\perp$  values. Successive integrations of electronic degrees of freedom on these shells from the (Fermi) energy cutoff  $E_F/k_B [= \pi t/(2k_B\sqrt{2})] \equiv 3000\text{K}$  down to zero at the Fermi surface result in the flow of the coupling constants toward their momentum dependent values as a function of temperature. This is carried out until a singularity is reached in the coupling constants which signals an instability of the electron gas against the formation of a broken symmetry state at a given temperature.

For the repulsive sector with this  $(\text{TMTSF})_2\text{X}$  model parameters, this can occur in either SDW or d-wave SC (SCd) channel depending on the amplitude of antinesting  $t'_\perp$ . The characteristic sequence of instabilities obtained for  $N_P = 60$  patches is reviewed in Figure 1<sup>36</sup>. At relatively low nesting deviations the magnetic scale  $T_{\text{SDW}}$  dominates; it drops with  $t'_\perp$  down to the critical value  $t'^*_\perp$  where instead of a plain quantum critical behaviour for antiferromagnetism for which  $T_{\text{SDW}}$  would reach zero, the ending of  $T_{\text{SDW}}$  gives rise to an SCd instability at its maximum  $T_c$ . The latter then steadily falls off with further increasing  $t'_\perp$ .

The normal phase we are interested in for the Seebeck coefficient is characterized by spin fluctuations. This is of course found in the SDW sector of the phase diagram where the SDW susceptibility  $\chi_{\text{SDW}}(\mathbf{q}_0)$  at the best nesting wave-vector of  $\epsilon^p_{\mathbf{k}}$  at  $\mathbf{q}_0 = (2k_F, \pi/d_\perp)$ , develops a singularity at  $T_{\text{SDW}}$ . In the SCd sector, an enhancement, though non singular, is still present. It takes the form of a Curie-Weiss temperature profile  $\chi_{\text{SDW}} \sim (T + \Theta)^{-1}$  over a large temperature domain above  $T_c$  ( $\Theta \geq 0$ ). The enhancement is quantum critical at  $t'^*_\perp$  ( $\Theta = 0$ ) and then decays with the decrease of  $T_c$  and the rise of the Curie-

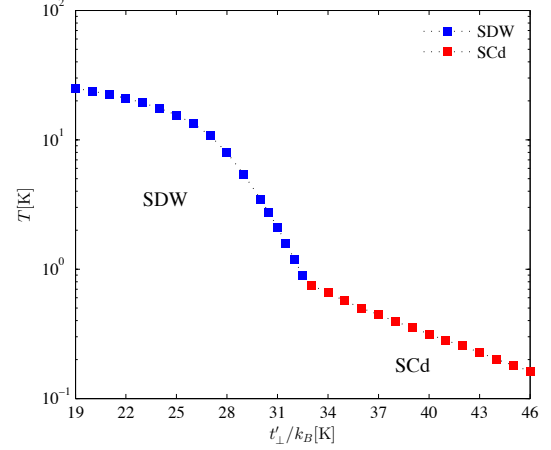


FIG. 1. Renormalization group results for the phase diagram of the quasi-1D electron gas model as a function of the antinesting tuning parameter  $t'_\perp$  and for the model parameters specified in Sec. III A.

Weiss scale  $\Theta$  along the antinesting axis<sup>33</sup>.

These short-range SDW correlations of the metallic phase are directly related to the enhancement of umklapp scattering entering the collision operator of the Boltzmann equation. In Figure 2, we show the temperature and momentum dependence of  $g_3$  on the Fermi surface, as projected in the  $(k_{\perp 1}, k_{\perp 3})$  plane when  $k_{\perp 1} = -k_{\perp 2}$  and  $k_{\perp 3} = -k_{\perp 4}$ . On the SDW side, the top panel of the Figure 2-a refers to the high temperature range ( $T > t_\perp$ ) which shows no structure in the transverse momentum plane for  $g_3$ , indicating that SDW correlations are essentially 1D in character and confined along chains. As temperature is lowered below  $t_\perp$ , transverse short-range order starts to develop, as shown by more intense scattering along the lines  $k_{\perp 1} = k_{\perp 3} \pm \pi$  ( $d_\perp = 1$ ). This is in accordance with the transverse momentum transfer associated with the best nesting wave-vector  $\mathbf{q}_0$  of the spectrum (15). When the lowest temperature is reached, peaks of stronger intensity appear on the corners at  $k_{\perp 1,3} = 0, \pm\pi$  and at the best nesting points  $\pm\pi/4, \pm 3\pi/4$  of the spectrum (15). These refer to warmer regions of scattering on the Fermi surface at the approach of the critical domain of the SDW instability.

On the SCd side of the phase diagram, in Fig. 2 (b), we see a pronounced but non singular anisotropic increase of umklapp scattering; peaks are confined around  $k_{\perp 1,3} = 0, \pm\pi$  on the Fermi surface, where enhanced scattering is found as the temperature is lowered. This enhancement occurs despite the flow of coupling constants towards a SCd fixed point indicative of a positive interference between both instabilities. This increase goes hand in hand with the one of SDW correlations in this temperature region, which are directly involved in the mechanism of d-wave Cooper pairing<sup>45–47</sup>. By increasing  $t'_\perp$  further, one can show that, although the same anisotropy of umklapp enhancement persists, its ampli-

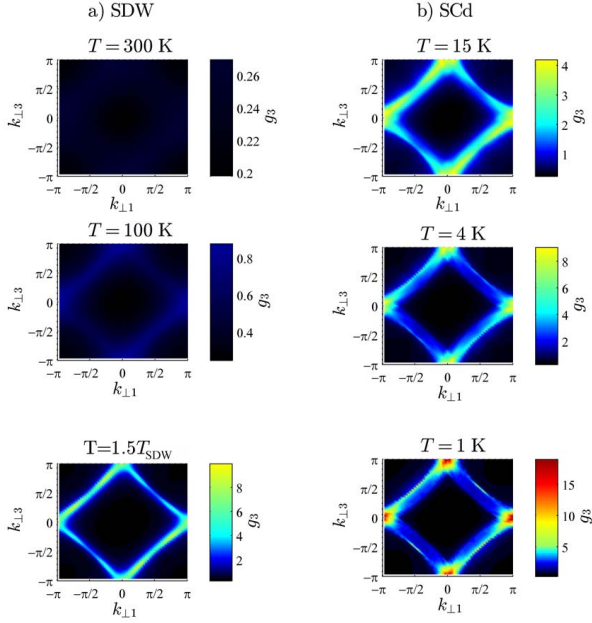


FIG. 2. Renormalized umklapp scattering amplitude  $g_3(k_{\perp 1}, -k_{\perp 1}; k_{\perp 3}, -k_{\perp 3})$ , projected in the  $(k_{\perp 1}, k_{\perp 3})$  plane at different temperatures for the metallic phase model parameters specified in Sec. III A. (a): SDW,  $t'_{\perp}/k_B = 25\text{K}$  ( $< t'^{*}_{\perp}/k_B$ ); (b) : SCd,  $t'_{\perp}/k_B = 35\text{K}$  ( $> t'^{*}_{\perp}/k_B$ ).

tude scales down with the reduction of  $T_c$ .

The consequence of this anisotropic growth of umklapp scattering on the temperature dependence of the Seebeck coefficient will be analyzed next.

#### IV. NUMERICAL RESULTS FOR THE SEEBECK COEFFICIENT

##### A. High temperature domain

The temperature dependence of the Seebeck coefficient (13), as obtained from the numerical solution of (A6) for the  $(\text{TMTSF})_2\text{X}$  model parameters, is shown in Fig. 3 (a) in the whole temperature interval of interest. By comparing the amplitude of the two contributions to the Seebeck coefficient in (13), we observe that apart from the high 1D temperature region the amplitude of the last term related to scattering dominates the band contribution  $Q_a^0$  (dashed line of Fig. 3 (a)) over most of the temperature interval. This gives rise to a shallow minimum for the Seebeck coefficient below which the normalized energy derivative  $\bar{\phi}'_{E=0, k_{\perp}} / \bar{\phi}_{E=0, k_{\perp}}$  of the normalized scattering time on the Fermi surface grows in importance, as meant in the lower panels of Fig. 4. The derivative is negative and according to (13), it gives a positive  $Q_a^c$ , as normally expected for holes carriers whose velocity and scattering time decrease with increasing energy.

By lowering temperature the scattering time deriva-

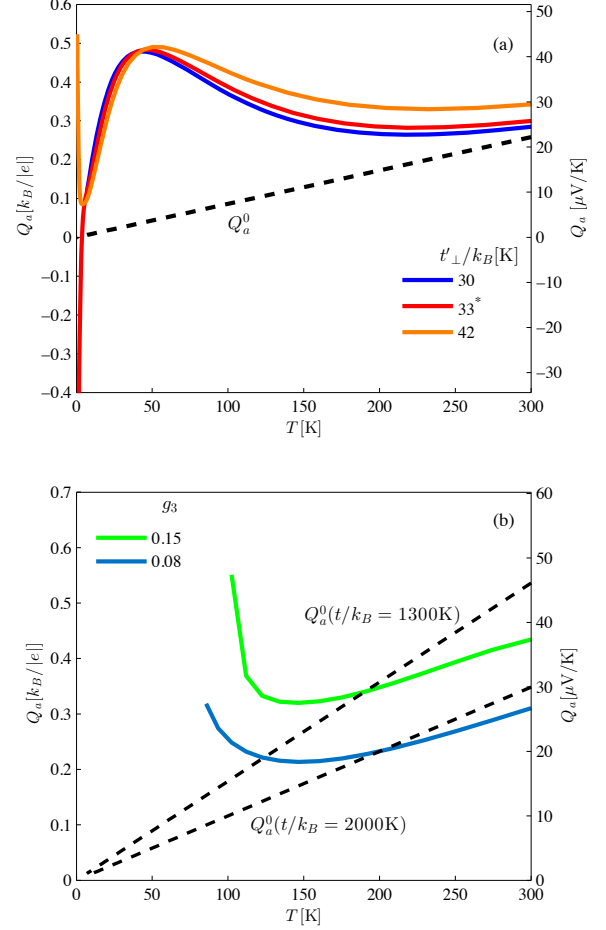


FIG. 3. The longitudinal Seebeck coefficient as a function of temperature for (a) different values of antinesting  $t'_{\perp}$  in the metallic phase and (b) for model parameters in the more correlated case with stronger umklapp scattering and lower amplitudes of hopping integrals ( $t'_{\perp}/k_B = 15\text{K}$ ,  $t_{\perp}/k_B = 100\text{K}$ , see Sec.V A on  $(\text{TMTTF})_2\text{X}$  salts). The dashed lines gives the band contribution  $Q_a^0$  of Eq. 13 for constant relaxation time in energy.

tive gains in amplitude and develops, like  $g_3$  of Fig. 2, anisotropy over the Fermi surface with maximums at  $k_{\perp} = 0, \pm\pi$  and  $\pm\pi/4, \pm 3\pi/4$ . This leads to a smooth increase of the Seebeck coefficient that levels off at a maximum value around the antinesting  $t'_{\perp}$  scale. This is followed in Fig. 3 (a) by a rapid drop at lower temperature which is nearly linear; it evolves toward anomalous features in the amplitude or the sign of the Seebeck coefficient depending on the distance to the critical value  $t'^{*}_{\perp}$  in the phase diagram. This will be discussed in more details below.

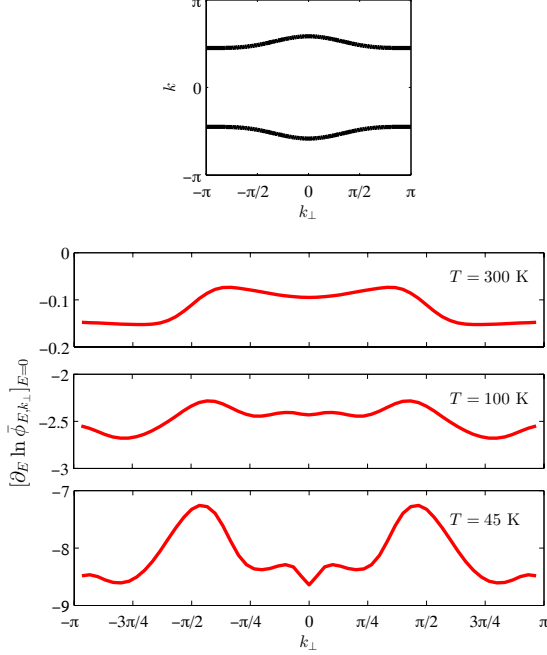


FIG. 4. Open Fermi surface of the quasi-1D electron gas model (top panel) and typical variations of the scattering time along the Fermi surface for different high temperatures ( $t'_\perp = t'^*_\perp$ ).

### B. Low temperature domain and quantum criticality

The results for the Seebeck coefficient in the metallic low temperature part of the phase diagram are presented in Fig. 5 (a) for different values of the antineesting parameter  $t'_\perp$ . On the SDW side, for  $t'_\perp$  relatively well below the critical  $t'^*_\perp/k_B$  ( $= 33$ K), the decrease of the Seebeck coefficient with lowering temperature is nearly linear as indicated by the constant ratio  $Q_a/T$  in Fig. 5 (b) when the temperature is lowered. Here the slope for  $Q_a$  is steeper than for the band contribution  $Q_a^0$  (dashed line Fig. 3 (a)). The dominant contribution to the Seebeck coefficient is coming from  $Q_a^c$  which is positive, resulting from a peak in the energy dependent quasi-particle scattering time located on the occupied side of the Fermi level at  $E < 0$ , as shown in the first top left panel of Figure 6 (a) above the SDW instability. It is worth noting that in these metallic conditions of the SDW state, the calculated scattering time at the Fermi level ( $\sim 10^{-9}$  sec) is significantly larger than the one found from a Drude theory of the conductivity of the Bechgaard salts above the SDW state<sup>48</sup>(see the note in Ref.<sup>49</sup>).

By raising  $t'_\perp$ , the temperature scale  $T_{\text{SDW}}$  in Figure 1 decays and at the approach of  $t'^*_\perp$  from below, the Seebeck coefficient develops an anomalous enhancement that is opposite in sign. This is depicted by the green lines

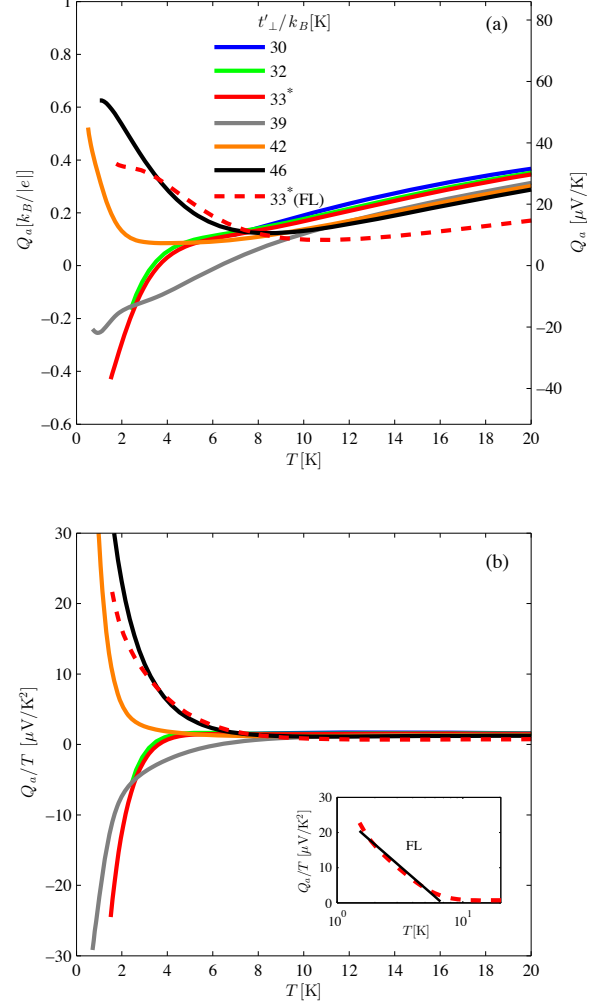


FIG. 5. (a) The longitudinal Seebeck coefficient  $Q_a$  and (b) the ratio  $Q_a/T$ , as a function of  $T$  at low temperature and different values of antineesting  $t'_\perp$ . The value with asterisk stands for the critical  $t'^*_\perp$ . The dashed line corresponds to the Fermi liquid limit using a momentum and temperature independent  $g_3 = 0.025$  at  $t'^*_\perp$ . The inset of part (b) displays the enhancement on a logarithmic temperature scale. The continuous line refers to  $Q_a/T \sim \ln T$ .

in Fig 5. The effect is reinforced when the electron system ultimately enters the SCd domain at  $t'^*_\perp$  where  $T_c$  is maximum. This indicates that the collision contribution  $Q_a^c$  is still negative or electron like in character and that it exceeds  $Q_a^0$  in amplitude. The sign reversal of the Seebeck coefficient refers to an increase of the scattering time with energy and then to a different asymmetry in the quasi-particle resonance peak of  $\langle \tilde{\phi}_{E,k_\perp} \rangle$ . According to Fig 6 (a), when the temperature is lowered, the latter is shifted from the occupied to the unoccupied side just above the Fermi level at  $E > 0$ . As for the anisotropy profile of  $\tilde{\phi}'/\tilde{\phi}$  over the Fermi surface, the third panel of Fig. 6 (b) reveals that this electron like component



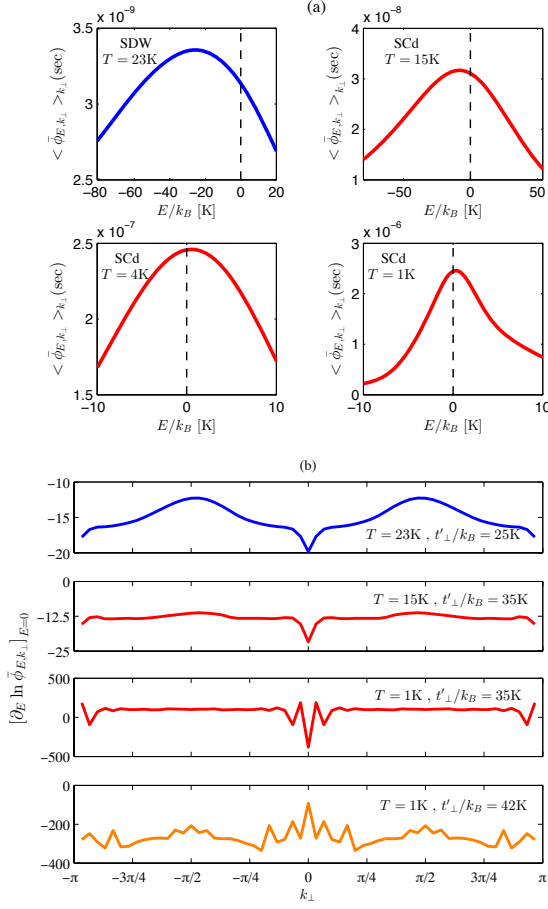


FIG. 6. (a) Variation of the normalized scattering time as a function of energy near the Fermi level at low temperature. Here  $t'_\perp/k_B = 25\text{K}$  and  $35\text{K}$  for the blue and red curves, respectively. (b) The anisotropy of the Seebeck coefficient along the Fermi surface at low temperature for different  $t'_\perp$ .

of the Seebeck coefficient comes in large part from the cold regions of scattering, namely, away from the warmer spots centered in  $k_\perp = 0$  and  $\pm\pi$  in the SCd sector [c.f. Fig. 2 (b)]. In the latter regions large oscillations of  $\tilde{\phi}'/\tilde{\phi}$  between positive and negative values tend to average out their contributions to a net positive contribution to the Seebeck coefficient. It is worth noting that the change of sign of the Seebeck coefficient, obtained by tuning  $t'_\perp$  across  $t'^*_\perp$ , occurs in the metallic state, that is in the absence of any reconstruction of the Fermi surface.

Further above  $t'^*_\perp$ , the negative enhancement of the Seebeck coefficient weakens and finally transforms into a positive upturn, as shown in Fig. 5. The latter is consistent with a quasi-particle resonance in the scattering time whose peak shifts back below the Fermi level with a negative slope in  $\langle \tilde{\phi}'_{E=0,k_\perp} \rangle_{k_\perp}$ , as shown in the lowest panel of Fig. 6 (b). This contributes positively to  $Q_a^c$ . At sufficiently large  $t'_\perp$ , namely when nesting alterations become large, the  $g_3$  coupling renormalizes less and less with a concomitant weaker anisotropy. In these conditions the Seebeck coefficient should tend to that of

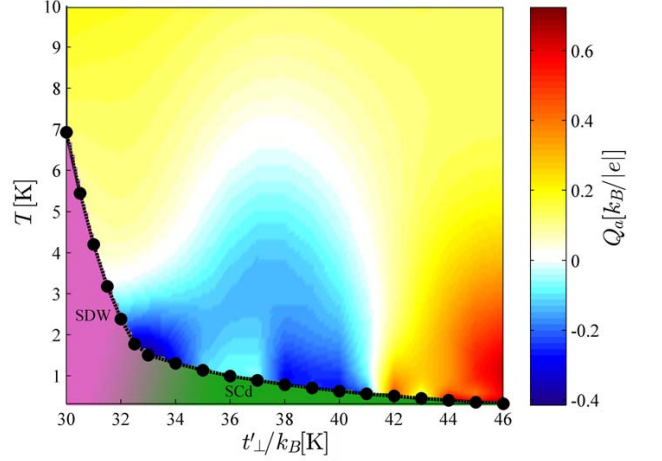


FIG. 7. Amplitude of the Seebeck coefficient at low temperature as a function of antineesting.

a Fermi liquid. This is confirmed when one imposes a momentum and temperature independent  $g_3$  in the calculations of the scattering time in (A6) which simulates the conditions of a Fermi liquid. This is shown by the dashed lines in Figure 5. It is worth noting that the Fermi liquid result for the quasi-1D electron gas model differs from the linear- $T$  band term  $Q_a^0$ . The corresponding ratio  $Q_a/T$  in Fig. 5 (b) displays a low temperature variation roughly congruent with a logarithmic enhancement. This has to be related to the fact with the fact that for a quasi-1D Fermi liquid, the scattering time is energy dependent and goes like  $\sim E^2 \ln E$  with logarithmic corrections; it is also asymmetric with respect to the Fermi level due to the presence of antineesting<sup>36,50</sup>. According to Figure 5 (b), the effect of renormalized umklapp leads to enhancements slightly to logarithmic corrections expected by previous predictions near a quantum critical point<sup>4</sup>.

Following the example of resistivity<sup>36</sup>, one can define from  $t'^*_\perp$  a characteristic zone of influence of quantum criticality where an anomalous sign of Seebeck coefficient is found. This is portrayed in Figure 7. As pointed out previously<sup>31,33,34,36</sup>,  $t'^*_\perp$  defines a quantum critical point where the entanglement or mutual reinforcement between SDW and SCd instabilities is the strongest,  $T_c$  the highest, and where spin fluctuations are quantum critical down to  $T_c$ <sup>33</sup>. This is apparently responsible for the electron type asymmetry in the energy dependence of electron-electron scattering time and therefore for the sign reversal of the Seebeck coefficient.

## V. COMPARISON WITH EXPERIMENTS IN LOW DIMENSIONAL ORGANIC CONDUCTORS

We now turn to the comparison of the above results with experiments. In this matter, it is instructive to first examine the Seebeck coefficient for some members of the (TMTTF)<sub>2</sub>X series, the so-called Fabre salts, which are known to exhibit a more correlated normal phase than the Bechgaard salts in normal pressure conditions as a result of stronger umklapp scattering<sup>23,51</sup>. The weak coupling RG can be used to compute the flow of umklapp scattering down to the approach of the Mott strong coupling region. After this incursion in the physics of the Fabre salts, we then proceed to the discussion of the Seebeck coefficient experiments in the Bechgaard salts in the light of the results of the present calculations.

### A. The Fabre salts (TMTTF)<sub>2</sub>X

The Fabre salts with  $X = \text{PF}_6, \text{AsF}_6, \text{Br}, \dots$ , form a series of quasi-1D conductors characterized by the same crystallographic structure as the Bechgaard salts (TMTSF)<sub>2</sub>X series<sup>52</sup>. The difference between the two series lies in the chemical composition of the TMTTF organic molecule for which the sulfur atoms are substituted in place of the selenium in TMTSF. In consequence the amplitude of the dimerization of TMTTF stacks turns out to be more pronounced in the solid state, along with band parameters that are typically smaller than those found in (TMTSF)<sub>2</sub>X (see Sec. III)<sup>44,53</sup>. In normal pressure conditions, the (TMTTF)<sub>2</sub>X are thus more one-dimensional in character and also more correlated than (TMTSF)<sub>2</sub>X through essentially a stronger influence of electronic umklapp processes.

This is exemplified by an upturn in electrical resistivity at the intermediate temperature,  $T_\rho$ , indicative of strong umklapp scattering that evolves towards an insulating 1D Mott behaviour<sup>51,54–56</sup>. Long-range ordered states can be found at much lower temperature which can involve charge, spin and even lattice degrees of freedom<sup>23</sup>. A remarkable property of the series emerges when sufficiently high pressure is applied to (TMTTF)<sub>2</sub>X which ultimately maps their physical properties to that of (TMTSF)<sub>2</sub>X at low pressure.

(TMTTF)<sub>2</sub>PF<sub>6</sub> is a prototype compound of the Fabre series characterized by the temperature scale  $T_\rho \simeq 220\text{K}$ ,<sup>54,55</sup>. The measurements of the Seebeck coefficient by Mortensen *et al.*,<sup>37</sup> for this compound are reproduced in Fig. 8 (a). The data show a monotonic increase of the Seebeck coefficient with decreasing temperature. The increase is consistent with non metallic behaviour shown by resistivity in the same range of temperature<sup>54,55</sup>.

The calculated results for a compound like (TMTTF)<sub>2</sub>PF<sub>6</sub> are displayed in Fig. 3 (b) when in accordance with band calculations<sup>44</sup>, smaller hopping terms ( $t/k_B = 1300\text{K}$ ,  $t_\perp/k_B = 100\text{K}$ ,  $E_F/k_B = 1500\text{K}$ ,  $t'_\perp/k_B = 15\text{K}$ )<sup>57</sup> and larger amplitudes for the bare

umklapp ( $g_3 = 0.15$ ) are used. With these figures, the instability at  $T_{\text{SDW}}$  occurs at much higher temperature ( $T_{\text{SDW}} = T_\rho \sim t_\perp$ ) and corresponds to the 1D Mott scale  $T_\rho$  at the one-loop level of the RG<sup>23,51</sup>. The important reduction of the longitudinal hopping  $t$  is responsible for a larger amplitude of the Seebeck coefficient, which is mainly dominated by the band term  $Q_a^0$  at high temperature, as shown by the dashed line of Fig. 3 (b). Note that this term surpasses the total  $Q_a$  indicating that the contribution of  $Q_a^c$  coming from collisions is relatively small but negative at very high temperature. The resulting  $Q_a$  then shows a smooth decrease with lowering temperature contrary to observation in (TMTTF)<sub>2</sub>PF<sub>6</sub>. However, the effect of the collision term becomes quickly positive and to give rise to an upturn of the Seebeck coefficient with lowering temperature as observed.

The case of (TMTTF)<sub>2</sub>Br is also of interest since along the pressure axis of a generalized phase diagram including both families, this compound is chemically shifted at about half distance between (TMTTF)<sub>2</sub>PF<sub>6</sub> and the members of (TMTSF)<sub>2</sub>X series at low pressure<sup>23</sup>. For normal state properties for instance, this is illustrated by the intermediate scale  $T_\rho \simeq 100\text{K}$  seen in resistivity<sup>54,58</sup>, in line with a smaller dimerization of the organic stacks for (TMTTF)<sub>2</sub>Br. The temperature variation of the Seebeck coefficient for the bromine salt is displayed in Fig. 8<sup>37</sup>. At room temperature the coefficient is smaller in amplitude compared to (TMTTF)<sub>2</sub>PF<sub>6</sub>; it drops as temperature is lowered, consistently with the more pronounced metallic character of this salt in this temperature range. However, the variation is not linear in temperature but reveals an enhancement with respect to the free carrier situation. Comes a minimum for temperature under  $T_\rho$ , followed by a rise that evolves toward a characteristic  $1/T$  behaviour for  $Q_a$  at sufficiently low temperature<sup>37</sup>, in congruence with a well defined insulating (Mott) gap.

By using intermediate figures for the band parameters ( $t = 2000\text{K}$ ,  $t_\perp = 100\text{K}$ ,  $E_F = 2200\text{K}$ ,  $t'_\perp/k_B = 15\text{K}$ ) and bare umklapp amplitude ( $g_3 = 0.08$ ), the amplitude of the calculated Seebeck coefficient at ambient temperature in Fig. 3 is intermediate between (TMTTF)<sub>2</sub>PF<sub>6</sub> and (TMTSF)<sub>2</sub>X, as shown in Fig.8 (a)-(b). The calculated decrease of  $Q_a$ , though enhanced compared to  $Q_a^0$  due to inelastic scattering, is less rapid than observed. The flow to strong umklapp scattering then results in the upturn in the Seebeck coefficient.

### B. The Bechgaard salts (TMTSF)<sub>2</sub>X

The Seebeck coefficient measured for the  $X=\text{PF}_6$  and  $\text{ClO}_4$  members of the Bechgaard is shown in Fig. 8 (b). Let us remind that in contrast with the sulfur based (TMTTF)<sub>2</sub>X compounds at low pressure,  $T_\rho$  is an irrelevant 1D scale for (TMTSF)<sub>2</sub>X, since these materials are metallic down to the temperature of onset of long-



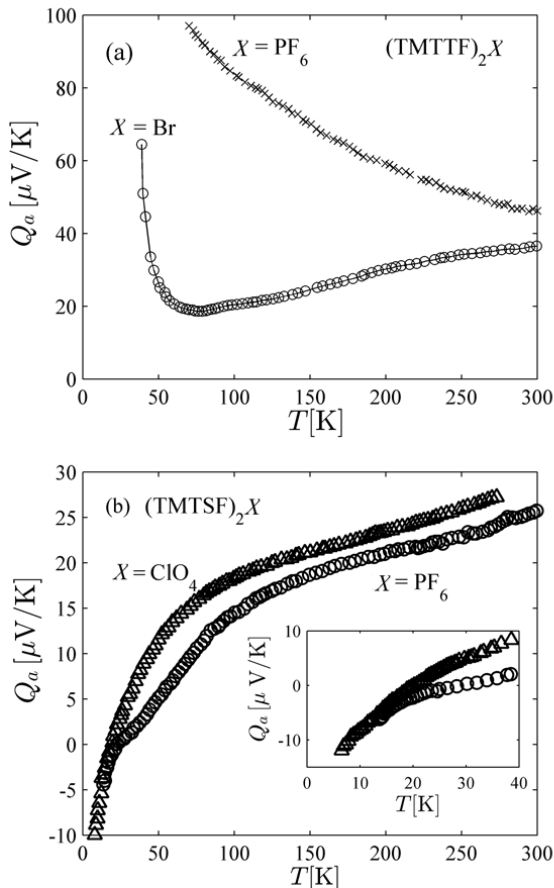


FIG. 8. Temperature dependence of Seebeck coefficients in (a): few (TMTTF)<sub>2</sub>X salts at ambient pressure (after Ref.<sup>37</sup>); (b): (TMTSF)<sub>2</sub>X in the metallic state above  $T_{\text{SDW}}$ ,  $X = \text{PF}_6$ , and  $T_c$ ,  $X = \text{ClO}_4$ . The inset shows the sign reversal of the Seebeck coefficient in low temperature domain (after Ref.<sup>15</sup>).

range order ( $T_{\text{SDW},c} \ll t_{\perp}$ ). In this range of temperature the system becomes effectively 2D regarding one-particle coherence, though strongly anisotropic. In Fig. 8 (b) we reproduced the temperature dependence of  $Q_a$  obtained by Chai *et al.*,<sup>15</sup> (see also Choi *et al.*,<sup>16</sup> Sun *et al.*,<sup>17</sup> Gubser *et al.*<sup>19</sup> and Chaikin<sup>18</sup>). The (TMTSF)<sub>2</sub>PF<sub>6</sub> shows SDW ordering at  $T_{\text{SDW}} \simeq 12.5\text{K}$ , whereas for (TMTSF)<sub>2</sub>ClO<sub>4</sub> the anion (ClO<sub>4</sub>) ordering in slow cooling conditions pushes the compound on the SC part of the phase diagram of series with  $T_c \simeq 1.2\text{K}$ <sup>22,59</sup>.

Close to ambient temperature, the Seebeck coefficient for both compounds reveals values relatively close to the calculated band limit  $Q_a^0$  given in Fig. 3 using the (TMTSF)<sub>2</sub>X band parameters of Sec. III. At lower temperature a positive enhancement with respect to a  $T$ -linear descent is observed for both compounds, in qualitative agreement with the one found in the present calculations in Fig. 3. However, in contrast to predictions, no maximum for  $Q_a$  is found experimentally; the data of Fig. 8 (b) rather show a shoulder like structure that precedes the low temperature descent of the Seebeck

coefficient. This suggests that the energy variation of the collision term is less rapid than predicted in this temperature range. It is not excluded, however, that such a maximum would show up if small pressure was applied to a compound like (TMTTF)<sub>2</sub>Br [see Fig. 8 (a)], which would suppress  $T_{\rho}$ <sup>58</sup> and shift the compound on the left hand side of (TMTSF)<sub>2</sub>X along the pressure axis<sup>23</sup> (see also the footnote in<sup>60</sup>).

For both compounds, the Fig. 8 (b) shows that the drop seen at low temperature for the Seebeck coefficient does not extrapolate toward zero, but exhibits negative enhancement from  $Q_a^c$  that evolves toward to a net sign reversal of the Seebeck coefficient. The fact that the  $T_c$  for (TMTSF)<sub>2</sub>ClO<sub>4</sub> is small, this sign reversal can be followed down to the lowest temperature of the metallic state, as shown in the insert of Fig. 8 (b). The sign reversal of the Seebeck coefficient occurs in the metallic state in the absence of a Fermi surface reconstruction that would transform the nature of carriers from hole to electron type. Therefore the present calculations provide an avenue of explanation for this effect in terms of an anomalous energy dependence of the inelastic umklapp scattering at the Fermi level that becomes electron like in character (see Fig. 6). This transformation takes its origin in the SDW fluctuations which act as the source of enhancement of umklapp scattering. As we have seen in Sec. III A, the development of these spin fluctuations can be greatly enhanced over sizable intervals of temperature and antinesting in the neighborhood of the quantum critical point  $t'_{\perp}^*$  [see for example Fig. 2 (b)]<sup>33</sup>, in qualitative correspondence with the spreading of the sign reversal of the Seebeck coefficient found in Figs. 5 and 7. It is worth stressing that NMR experiments for the temperature variation of the nuclear spin lattice relaxation rate have brought considerable evidence for the presence of spin fluctuations for both compounds in the same temperature range and their amplitude with pressure<sup>25,28,29,41,61,62</sup>.

## VI. SUMMARY AND CONCLUDING REMARKS

In the work developed above, a derivation of the Seebeck coefficient in quasi-1D interacting electron systems has been carried out from a numerical solution of the linearized Boltzmann equation using the renormalization group method for the evaluation of the electron-electron scattering matrix element. From a parametrization of the electron gas model compatible with the spin-density-wave to superconducting sequence of orderings found in organic superconductors under pressure, the temperature variation of the Seebeck coefficient in the metallic phase was calculated. It was shown to develop marked deviations with respect to the hole band linear- $T$  prediction. These deviations found their striking expression in the quantum critical region of the metallic phase linked to the juncture of antiferromagnetic and superconducting orders. It is where the enhancement of the

Seebeck coefficient undergoes a sign reversal, attributable to an anomalous low energy variation of the anisotropic electron-electron scattering time becoming electron-like in character over most of the Fermi surface. Spin fluctuations, which act as a source of inelastic umklapp scattering, appear as a key determinant for this sign reversal which occurs in the absence of a Fermi surface reconstruction. It is only when the antineesting parameter, which simulates the role of pressure in real materials like the organics, is tuned sufficiently far away from the quantum critical point that a Fermi liquid type of enhancement of the Seebeck coefficient is recovered.

The results were shown to capture many features of existing data in quasi-1D conductors like the Bechgaard salts  $(\text{TMTSF})_2X$ , in particular the crossover to negative values of the Seebeck coefficient at low temperature in the neighbourhood of their quantum critical point along the pressure axis. The size of the enhancement with respect to the band prediction is also fairly well taken into account, suggesting that electron-electron scattering in the presence of electron-hole asymmetry mainly due to nesting alterations is likely to be the most important processes governing the temperature dependence of the thermoelectric response of these materials. The results of the calculations being obtained at arbitrary antineesting distance from the quantum critical point of the phase diagram, can serve as a stimulus for future experiments of the Seebeck coefficient in Bechgaard salts as function of applied hydrostatic pressure. Such a systematic study is lacking so far. Following the example of previous electrical transport<sup>27,63</sup> and NMR studies<sup>25,28,29,64</sup>, it would be worthwhile to check if the Fermi liquid behaviour for the Seebeck coefficient is recovered sufficiently far above the quantum critical pressure.

The present theory of the Seebeck coefficient was also applied to the more correlated Fabre  $(\text{TMTTF})_2X$  series, whose members with centro-symmetrical anions  $X$  are known to become Mott insulators at relatively high temperature in normal pressure conditions. Stronger umklapp scattering and narrower bands characterize these materials. This precipitates an instability toward an insulating state at much higher temperature and accordingly, yields a pronounced enhancement of the Seebeck coefficient that is present in experiments.

## ACKNOWLEDGMENTS

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### Appendix A: Linearized Boltzmann equation

In the presence of an external longitudinal thermal gradient and induced electric field, the linearized Boltzmann equation for the normalized deviations  $\bar{\phi}_{\mathbf{k}}$  ( $= \bar{\phi}_{\mathbf{k}}^{j=\mathcal{E},T}$ ) can be put in the single form of Eq. (14):

$$\mathcal{L}\bar{\phi}_{\mathbf{k}} = \sum_{i=1}^4 \sum_{\mathbf{k}'} \mathcal{L}_{\mathbf{k},\mathbf{k}'}^{[i]} \bar{\phi}_{\mathbf{k}'} = 1, \quad (\text{A1})$$

The collision operator is expressed as the sum of four terms,

$$\begin{aligned} \sum_{i=1}^4 \mathcal{L}_{\mathbf{k},\mathbf{k}'}^{[i]} &= \frac{1}{(LN_P)^2} \sum_{\mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4} \frac{1}{2} |\langle \mathbf{k}, \mathbf{k}_2 | g_3 | \mathbf{k}_3, \mathbf{k}_4 \rangle - \langle \mathbf{k}, \mathbf{k}_2 | g_3 | \mathbf{k}_4, \mathbf{k}_3 \rangle|^2 \times \frac{2\pi}{\hbar} \delta_{\mathbf{k}+\mathbf{k}_2, \mathbf{k}_3+\mathbf{k}_4+p\mathbf{G}} \\ &\times \delta(\varepsilon_{\mathbf{k}}^p + \varepsilon_{\mathbf{k}_2}^{p_2} - \varepsilon_{\mathbf{k}_3}^{p_3} - \varepsilon_{\mathbf{k}_4}^{p_4}) \frac{f^0(\mathbf{k}_2)[1 - f^0(\mathbf{k}_3)][1 - f^0(\mathbf{k}_4)]}{[1 - f^0(\mathbf{k})]} (\delta_{\mathbf{k},\mathbf{k}'} + \delta_{\mathbf{k}_2,\mathbf{k}'} - \delta_{\mathbf{k}_3,\mathbf{k}'} - \delta_{\mathbf{k}_4,\mathbf{k}'}). \end{aligned} \quad (\text{A2})$$

The amplitude of umklapp vertex are evaluated by the RG in the framework of the quasi-1D electron gas model,

$$\begin{aligned} \langle \mathbf{k}_1, \mathbf{k}_2 | g_3 | \mathbf{k}_3, \mathbf{k}_4 \rangle &= \pi \hbar v_F g_3(\mathbf{k}_{F,1}^p, \mathbf{k}_{F,2}^p; \mathbf{k}_{F,3}^{-p}, \mathbf{k}_{F,4}^{-p}) \\ &= \pi \hbar v_F g_3(k_{\perp 1}, k_{\perp 2}; k_{\perp 3}, k_{\perp 4}), \end{aligned} \quad (\text{A3})$$

where the position on the Fermi surface is parametrized by the transverse wavevectors. To solve the equation with the explicit form of matrix elements shown in equation A2, we separate the momentum conservation con-

straint into longitudinal and transverse components,<sup>50</sup>

$$\begin{aligned} \delta_{\mathbf{k}+\mathbf{k}_2, \mathbf{k}_3+\mathbf{k}_4+p\mathbf{G}} &= \delta_{k_{\perp}+k_{\perp 2}, k_{\perp 3}+k_{\perp 4}} \\ &\times \frac{2\pi}{L} \hbar v_F \delta(\varepsilon_{\mathbf{k}}^p + \varepsilon_{\mathbf{k}_2}^{p_2} + \varepsilon_{\mathbf{k}_3}^{p_3} + \varepsilon_{\mathbf{k}_4}^{p_4} - \Sigma), \end{aligned} \quad (\text{A4})$$

where  $\Sigma = \epsilon_{\perp}(k_{\perp}) + \epsilon_{\perp}(k_{\perp 2}) + \epsilon_{\perp}(k_{\perp 3}) + \epsilon_{\perp}(k_{\perp 4})$ . The summation over the momentum vectors can be written as

$$\frac{1}{LN_P} \sum_{\mathbf{k}} = \sum_p \int \frac{d\varepsilon_{\mathbf{k}}^p}{2\pi \hbar v_F} \frac{1}{N_P} \sum_{k_{\perp}}. \quad (\text{A5})$$

Carrying out the integration over  $\varepsilon_{\mathbf{k}'}^{p'}$  and by rearranging the terms, we arrive at the final equation,

$$\begin{aligned} & \frac{\pi}{\beta\hbar} \frac{1}{N_P^2} \sum_{k'_\perp, k_{\perp 3}, k_{\perp 4}} \left\{ |g_3(k_\perp, k_{\perp 3} + k_{\perp 4} - k_\perp; k_{\perp 3}, k_{\perp 4}) - g_3(k_\perp, k_{\perp 3} + k_{\perp 4} - k_\perp; k_{\perp 4}, k_{\perp 3})|^2 \delta_{k_\perp, k'_\perp} \right. \\ & \times \frac{\beta\Sigma'/4 \cosh(\beta E/2)}{\cosh(\beta(\Sigma'/4 - E/2)) \sinh(\beta\Sigma'/4)} + |g_3(k_\perp, k'_\perp; k_{\perp 3}, k_{\perp 4}) - g_3(k_\perp, k'_\perp; k_{\perp 4}, k_{\perp 3})|^2 \\ & \times \frac{\beta\Sigma''/4 \cosh(\beta E/2)}{\cosh(\beta(\Sigma''/4 - E/2)) \sinh(\beta\Sigma''/4)} \delta_{k_\perp + k'_\perp, k_{\perp 3} + k_{\perp 4}} - 2|g_3(k_\perp, k_{\perp 3}; k'_\perp, k_{\perp 4}) - g_3(k_\perp, k_{\perp 3}; k_{\perp 4}, k'_\perp)|^2 \\ & \times \left. \frac{\beta\Sigma''/4 \cosh(\beta E/2)}{\cosh(\beta(\Sigma''/4 - E/2)) \sinh(\beta\Sigma''/4)} \delta_{k_\perp + k_{\perp 3}, k'_\perp + k_{\perp 4}} \right\} \bar{\phi}_{E, k'_\perp} = 1, \end{aligned} \quad (\text{A6})$$

where  $\Sigma' = \epsilon_\perp(k_\perp) + \epsilon_\perp(k_{\perp 3} + k_{\perp 4} - k_\perp) + \epsilon_\perp(k_{\perp 3}) + \epsilon_\perp(k_{\perp 4})$  and  $\Sigma'' = \epsilon_\perp(k_\perp) + \epsilon_\perp(k'_\perp) + \epsilon_\perp(k_{\perp 3}) + \epsilon_\perp(k_{\perp 4})$ . By inserting the RG results of Sec. III A for the momen-

tum resolved umklapp scattering, the numerical solution of (A6) for  $\bar{\phi}_{E, k_\perp}$  allows the evaluation of the scattering contribution  $Q_a^c$  to the Seebeck coefficient (13).

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